Supplementary Information

Syntheses and Electrochemical Properties of $(Nb_{1-y}Ti_y)_4C_3T_x$ Solid Solution MXenes

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In the preparation of MAX phase precursors, the atomic ratio of Nb: Ti: Al: C is maintained at 4-4y:4y:1.5:2.7, where y takes values of 0.1, 0.2, and 0.3. Under these conditions, it is possible to obtain 413 phases without 312 phase impurities. However, when the y value is 0.4, a mixture of the 413 and 312 MAX phases is formed, as shown in Fig. S1. For y values of 0.6 and 0.8, only the 312 phase is obtained. Similar results are observed when 5 at% TiO_2 (Fig. S2) or 5 at% Nb_2O_5 (Fig. S3) is added to the precursors.

The pure 413 MAX phase, $(Nb_{0.6}Ti_{0.4})_4AlC_3$ was successfully synthesized with the atomic ratio of Nb : Nb_2O_5 : Ti : Al : C equal to 2.85 : 0.075 : 2 : 1.2 : 3.5, as shown in Fig. 1a. Furthermore, when $y = 0.6$ or 0.8, regardless of the ratios, pure 413 MAX phase cannot be formed.[1]

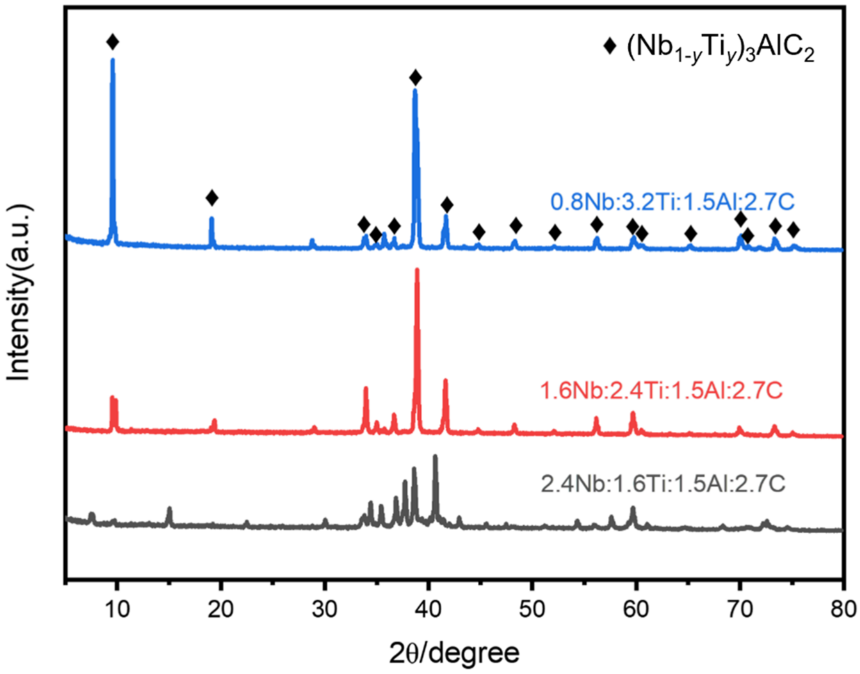

Fig. S1 XRD spectra of MAX phase materials obtained with varying Ti/(Nb+Ti) ratios without oxide).

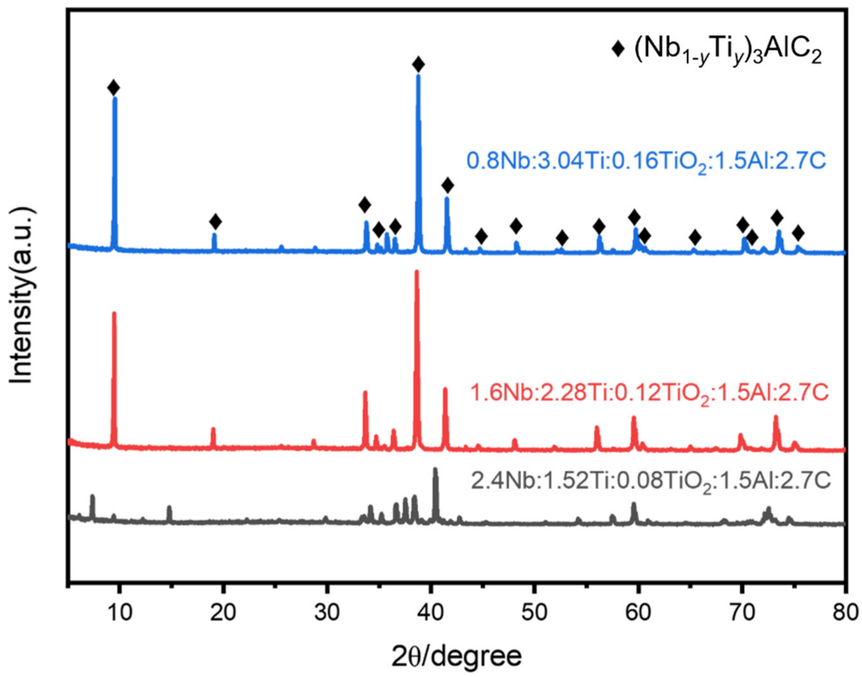

Fig. S2 XRD spectra of MAX phase materials obtained with varying Ti/(Nb+Ti) ratios, where 5at% of Ti was replaced with TiO_2.

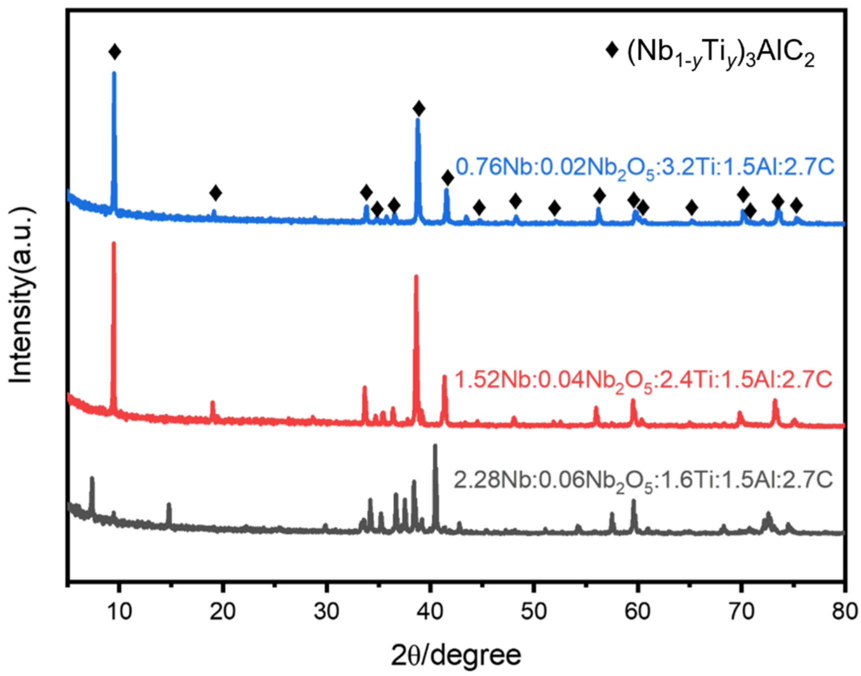

Fig. S3 XRD spectra of MAX phase materials obtained with varying Ti/(Nb+Ti) ratios, where 5at% of Nb was replaced with Nb_2O_5.

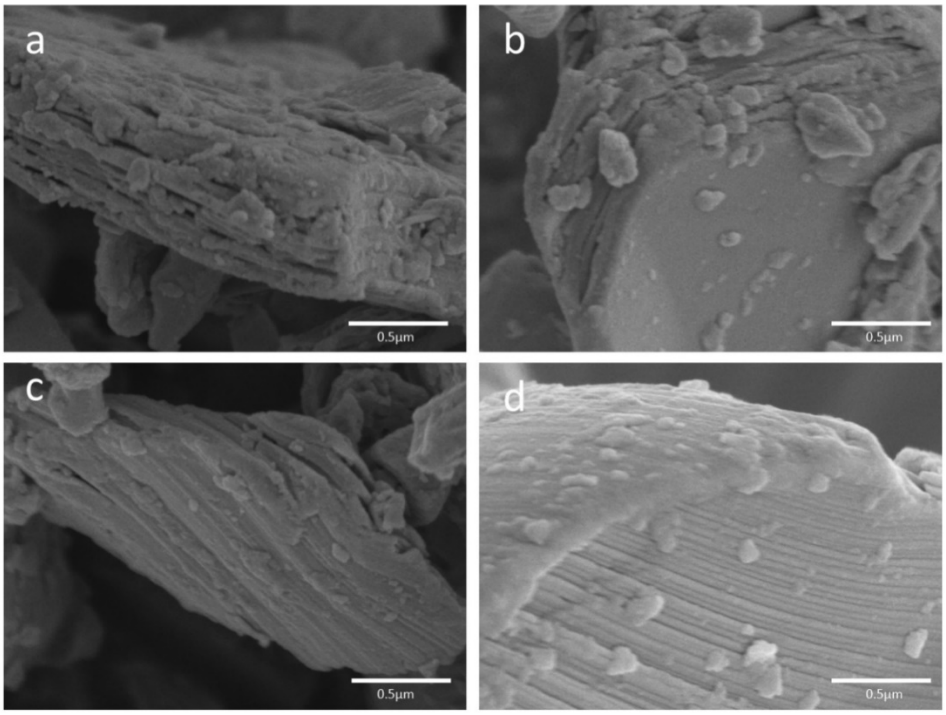

Fig. S4 SEM images of (a) Nb_4AlC_3, (b) $(Nb_{0.9}Ti_{0.1})_4AlC_3$, (c) $(Nb_{0.8}Ti_{0.2})_4AlC_3$ and (d) $(Nb_{0.6}Ti_{0.4})_4AlC_3$

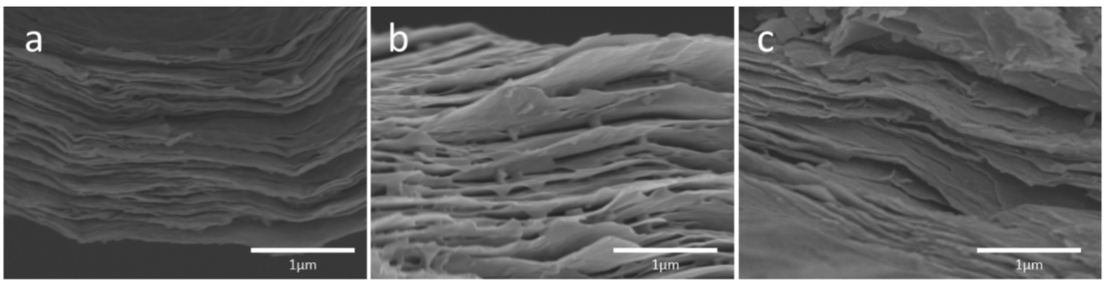

Fig. S5 SEM cross section view of (a) $Nb_4C_3T_x$, (b) $(Nb_{0.9}Ti_{0.1})_4C_3T_x$ and (c) $(Nb_{0.8}Ti_{0.2})_4C_3T_x$.

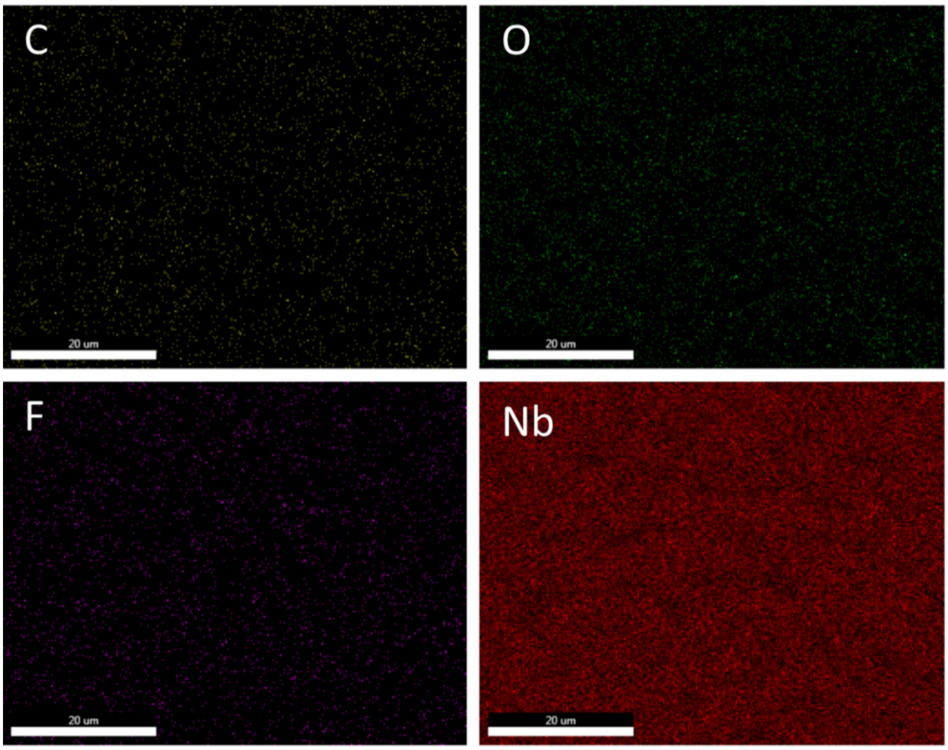

Fig. S5 The EDS element mapping of $Nb_4C_3T_x$.

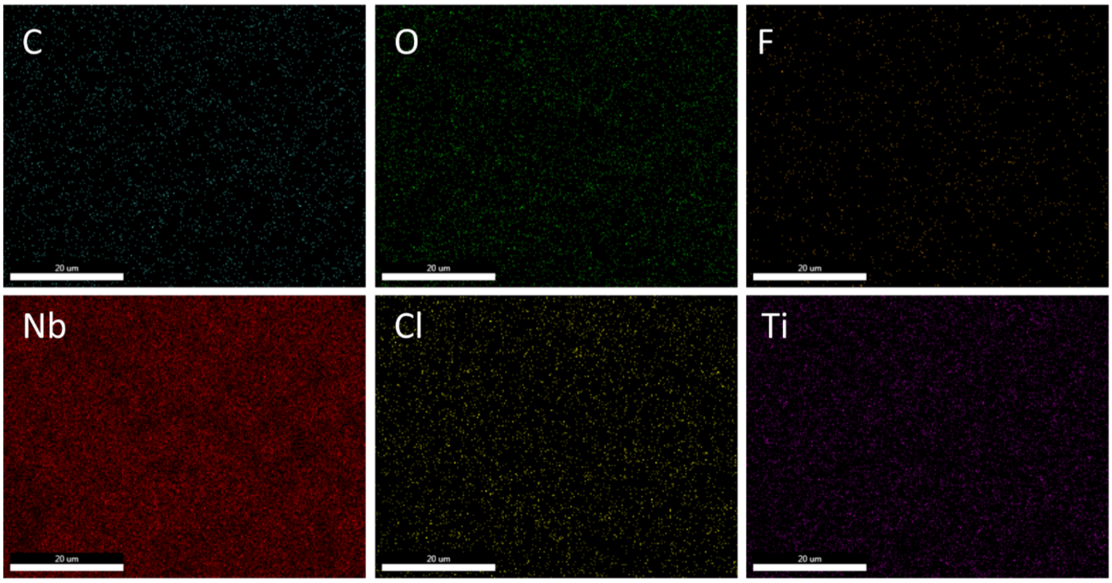

Fig. S7 The EDS element mapping of $(Nb_{0.9}Ti_{0.1})_4C_3T_x$.

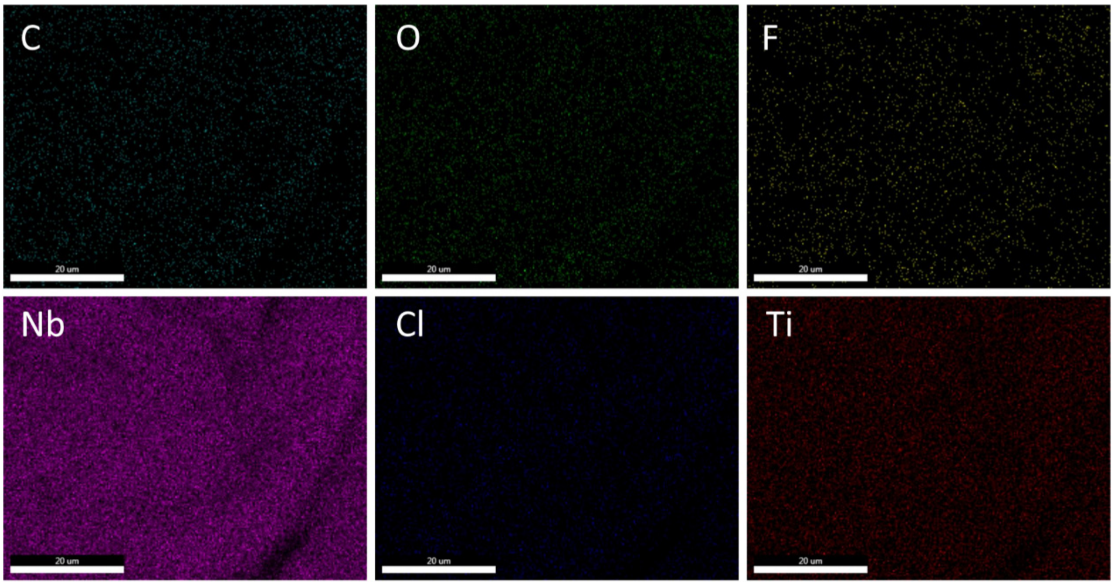

Fig. S8 The EDS element mapping of $(Nb_{0.8}Ti_{0.2})_4C_3T_x$.

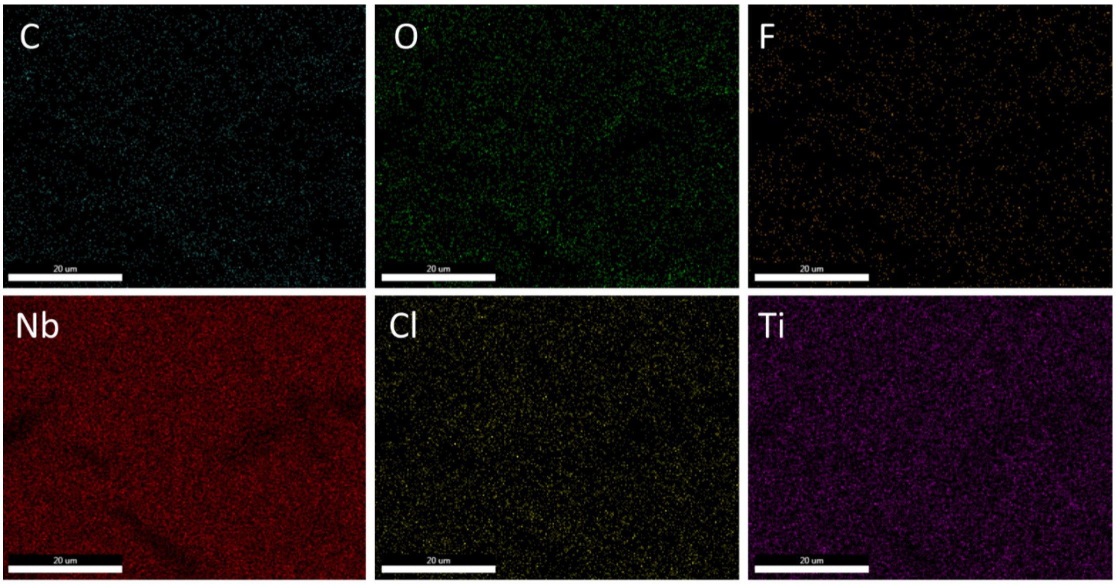

Fig. S9 The EDS element mapping of $(Nb_{0.7}Ti_{0.3})_4C_3T_x$.

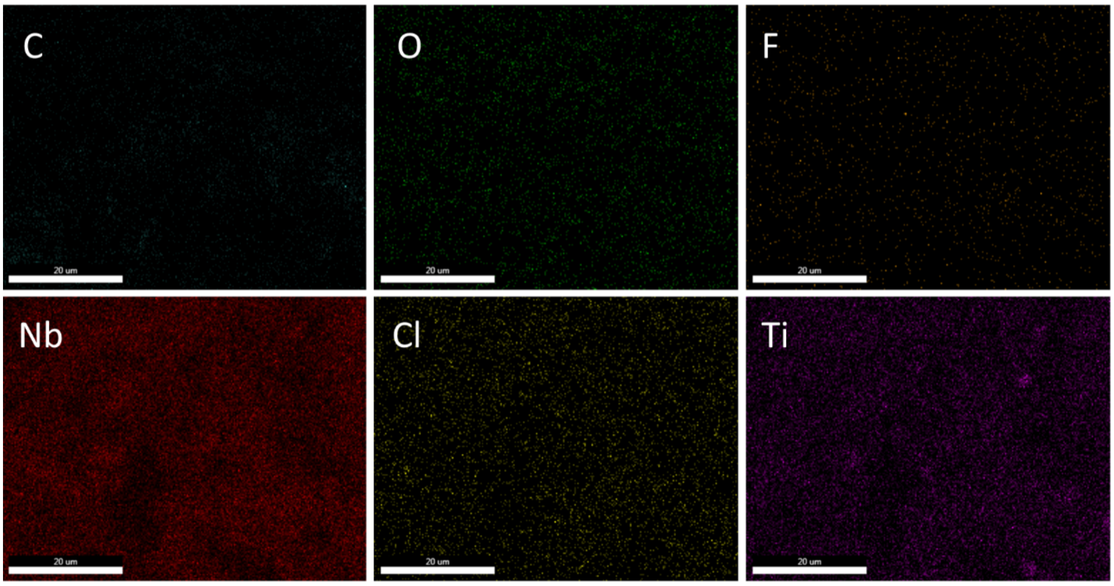

Fig. S10 The EDS element mapping of $(Nb_{0.6}Ti_{0.4})_4C_3T_x$.

Table S1 Mass load of $(Nb_{1-y}Ti_y)_4C_3T_x$ for the working electrode

	Quality (mg)	Area(cm^2)
$Nb_4C_3T_x$	0.294	0.283
$(Nb_{0.9}Ti_{0.1})_4C_3T_x$	0.705	0.283
$(Nb_{0.8}Ti_{0.2})_4C_3T_x$	0.270	0.2827
$(Nb_{0.7}Ti_{0.3})_4C_3T_x$	0.290	0.283
$(Nb_{0.6}Ti_{0.4})_4C_3T_x$	0.270	0.283

Table S2 The elemental composition of $(Nb_{1-y}Ti_y)_4C_3T_x$ MXenes based on EDS analysis

	Nb	Ti	C	O	F	Cl	Ti/(Nb+Ti)
$Nb_4C_3T_x$	33.98	-	42.73	22.02	1.28	-	-
$(Nb_{0.9}Ti_{0.1})_4C_3T_x$	32.22	3.79	39.56	23.85	0.00	0.63	10.52%
$(Nb_{0.8}Ti_{0.2})_4C_3T_x$	33.8	8.78	34.85	20.82	0.55	1.21	20.62%
$(Nb_{0.7}Ti_{0.3})_4C_3T_x$	21.75	10.50	41.58	23.75	0.77	1.65	32.56%
$(Nb_{0.6}Ti_{0.4})_4C_3T_x$	10.62	8.75	59.69	17.83	1.92	1.2	45.17%

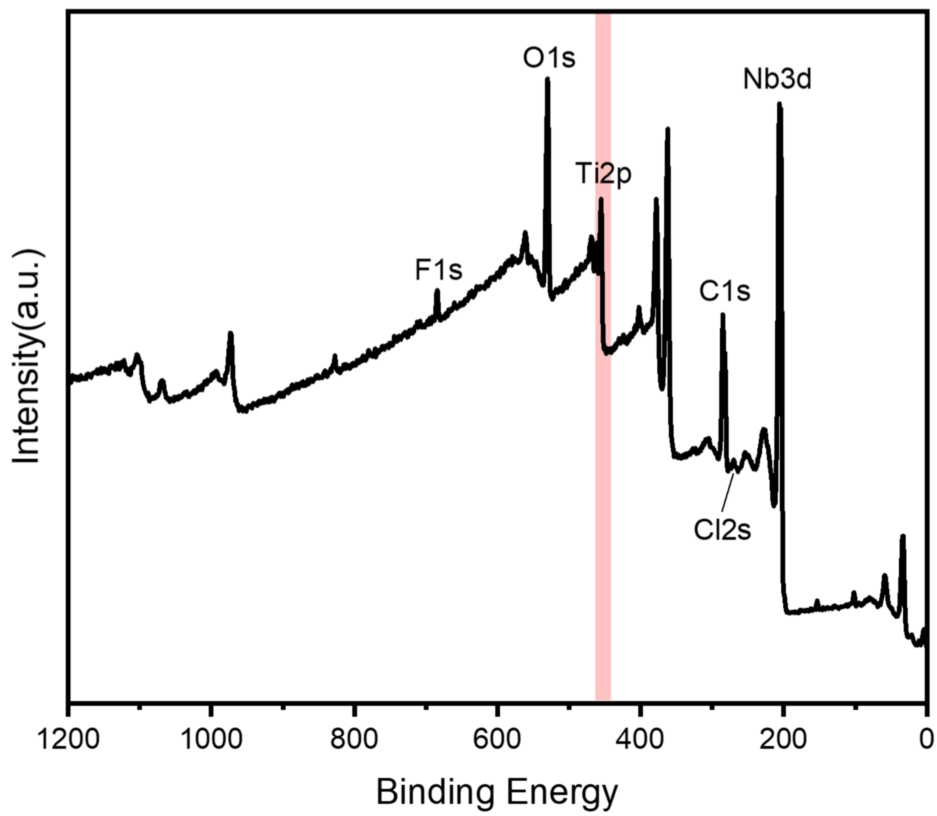

Fig. S11 XPS full spectrum of $(Nb_{0.7}Ti_{0.3})_4C_3T_x$.

Table S3 The elemental composition of $(Nb_{0.7}Ti_{0.3})_4C_3T_x$ based on XPS analysis.

	Nb	Ti	C	O	F	Cl	Ti/(Nb+Ti)
$(Nb_{0.7}Ti_{0.3})_4C_3T_x$	22.12	8.87	24.98	24.67	16.67	2.68	28.62%

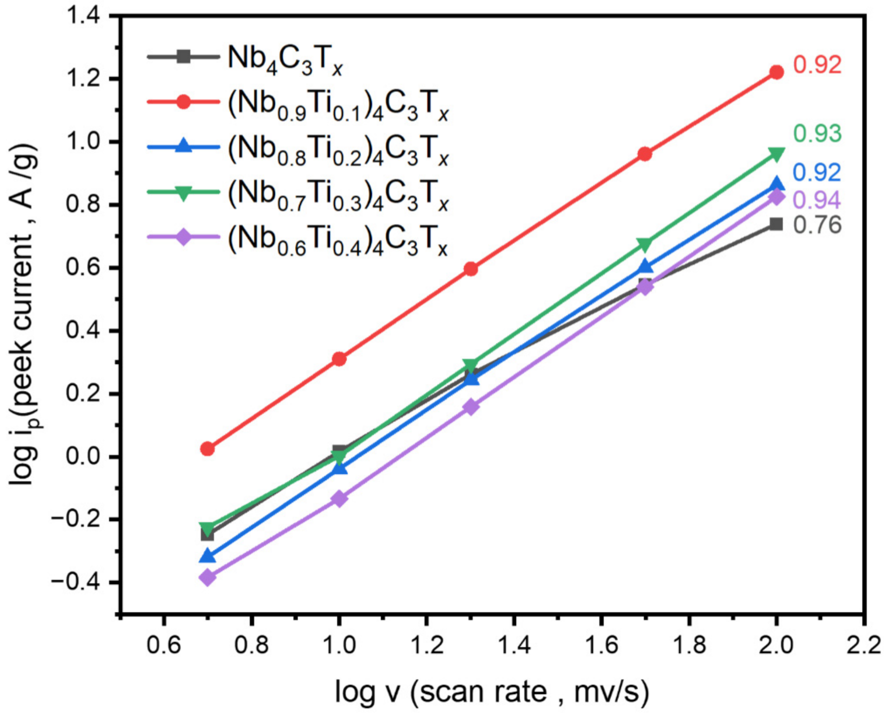

Fig. S12 b-value determination by plotting log (specific current) versus log (scan rate) and the current and scan rate was taken from CV profiles in Fig. 4(a~e).

References

1. M. Downes, C. E. Shuck, R. W. Lord, M. Anayee, M. Shekhirev, R. J. Wang, T. Hryhorchuk, M. Dahlqvist, J. Rosen and Y. Gogotsi, *Acs Nano*, 2023, **17**, 17158-17168.